



Proposed Amendment for:  
10/596,750 (789-100)  
Attachment to Interview Summary  
Do not enter claims

## Patent Technology Centers

### Facsimile Transmission

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### Fax Notes:

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Mr. Byrne,

The following is a proposed amendment to put 10/596,750 in condition for allowance. Included are a marked-up version, a clean version, and comments. The comments identify support in the specification for the property limitations set forth in claims 10, 20, 26, 27, 33, and 35. If acceptable, I will enter these changes in an examiner's amendment.

Sincerely,

Michael J Feely (Primary Examiner; AU 1761)

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***Marked-up Version***

1. (Proposed Amendment) A method for making a modified epoxy, comprising the steps of:

a) preparing a clay solution comprising solvents, clay particles of a dimension in the nanometer range and agglomerates of clay particles;

b) generating a flow of clay solution and submitting said flow to: (1) high pressure to generate high velocity and to allow shearing in the clay solution to occur; (2) a region of obstacles allowing the agglomerates of clay particles to be broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and

c) mixing the dispersed clay solution with at least a pristine epoxy, wherein a rubber material is optionally provided with the pristine epoxy during mixing.

2. (Cancelled)

3. (Proposed Amendment) The method according to claim 1, wherein said step a) comprises incorporating a first part of the pristine epoxy into the clay solution; and said step c) comprises mixing the dispersed clay ~~particles~~ solution with a remaining part of the pristine epoxy.

4. (Original) The method according to claim 1, wherein said step a) comprises mixing with at least one of mechanical and ultrasonic mixing.

5. (Original) The method according to claim 1, wherein said step b) comprises submitting the clay solution to a pressure of about 20,000 psi in tubes of a diameter of about 0.1 mm.
6. (Original) The method according to claim 1, wherein said step b) comprises exfoliating the clay particles in the solution.
7. (Previously Presented) The method according to claim 1, wherein said step c) comprises adding curing agents to yield a solid epoxy material.
8. (Proposed Amendment) The method according to claim 1, whereby the modified epoxy comprises agglomerates of less than about 1  $\mu\text{m}$  and agglomerates of a maximum diameter ~~comprised~~ between about 1  $\mu\text{m}$  and 2  $\mu\text{m}$ .
9. (Previously Presented) The method according to claim 1, whereby the modified epoxy has enhanced viscoelastic properties and improved fracture toughness compared to the pristine epoxy.
10. (Proposed Amendment) The method according to claim 9, wherein a content of about 1 wt% of clay loading and no optional rubber yields ~~whereby the modified epoxy has~~ an increase in critical stress intensity ( $K_{1C}$ ) and critical strain energy release rate ( $G_{1C}$ ) of up to 2 and 3 times ~~respectively, with respect to the pristine epoxy, at about 1 wt % of clay loading.~~

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11. (Original) The method according to claim 1, whereby the modified epoxy has enhanced barrier properties, including water absorption resistance, adhesion strength and flammability resistance, with respect to the pristine epoxy.

12. (Original) The method according to claim 1, wherein a mixture of clay and epoxy obtained has a stability over an extended period of time.

13. (Original) The method according to claim 1, wherein the optional rubber is provided with the pristine epoxy during mixing ~~is a rubber modified epoxy resin.~~

14 - 19. (Cancelled)

20. (Proposed Amendment) A modified epoxy produced from a pristine epoxy, the modified epoxy having at least higher barrier properties and thermal resistance, a higher critical stress intensity factor ( $K_{IC}$ ), and a higher critical strain energy release rate ( $G_{IC}$ ) than the pristine epoxy, the modified epoxy produced by:

a) preparing a clay solution comprising solvents, clay particles of a dimension in the nanometer range and agglomerates of clay particles;

b) generating a flow of clay solution and submitting said flow to: (1) high pressure; (2) high velocity and breaking impacts in a region of obstacles to allow the agglomerates to be broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and

c) mixing the dispersed clay solution with at least part of the pristine epoxy, wherein a rubber material is optionally provided with the pristine epoxy during mixing;

wherein a content of about 1wt% of clay loading and no optional rubber yields an increase in  $K_{IC}$  and  $G_{IC}$  of ~~up to~~ 2 and 3 times respectively, with respect to the pristine epoxy.

21. (Original) The modified epoxy according to claim 20, comprising finely dispersed clay agglomerates of less than about 1  $\mu\text{m}$  and agglomerates of a maximum diameter between about 1  $\mu\text{m}$  and 2  $\mu\text{m}$ .

22. (Cancelled)

23. (Previously Presented) The modified epoxy according to claim 21, wherein the optional rubber is provided with the pristine epoxy during mixing.

24. (Original) The modified epoxy according to claim 21, further comprising additives.

25. (Cancelled)

26. (Proposed Amendment) The modified epoxy according to claim 23, wherein the optional rubber is CTBN, and a content of 6 phr of clay loading and 20 phr of CTBN yields an increase in  $K_{IC}$  and  $G_{IC}$  of ~~up to~~ 2.2 and 7.6 times respectively, with respect to the pristine epoxy.

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27. (Proposed Amendment) The method according to claim 13, wherein the optional rubber is CTBN, and a content of 6 phr of clay loading and 20 phr of CTBN yields modified epoxy has an increase in critical stress intensity factor ( $K_{IC}$ ) and critical strain energy release rate ( $G_{IC}$ ) of ~~up to~~ 2.2 and 7.6 times respectively, with respect to at 6-phr organoclay loading and 20-phr CTBN ~~compared with~~ the pristine epoxy.

28. (Proposed Cancellation)

29. (Proposed Cancellation)

30. (Proposed Cancellation)

31. (Proposed Cancellation)

32. (Proposed Cancellation)

33. (Proposed Amendment) A modified epoxy produced from a pristine epoxy, the modified epoxy having at least a higher flammability resistance and a higher fracture toughness than the pristine epoxy, the modified epoxy produced by:

- a) preparing a clay solution comprising ~~comprising~~ solvents, clay particles of a dimension in the nanometer range and agglomerates of clay particles;
- b) generating a flow of clay solution and submitting said flow to: (1) high pressure; (2) high velocity and breaking impacts in a region of obstacles to allow the agglomerates to be

broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and

c) mixing the dispersed clay solution with at least part of the pristine epoxy;

wherein a content of 6 ~~as low as about 4~~ phr of clay loading yields an increase in critical strain energy release rate ( $G_{IC}$ ) of ~~at least 4.8~~ 5.8 times, with respect to the pristine epoxy.

34. (Proposed Amendment) The modified epoxy of claim 33, wherein said step c) comprises adding curing agents to yield a solid epoxy material.

35. (Proposed Amendment). A modified epoxy produced from a rubber-modified pristine epoxy, the modified epoxy having at least a higher flammability resistance and a higher fracture toughness than the rubber-modified pristine epoxy, the modified epoxy produced by:

a) ~~forming~~ preparing a clay solution comprising ~~comprising solvents~~, clay particles of a dimension in the nanometer range and agglomerates of clay particles;

b) generating a flow of clay solution and submitting said flow to: (1) high pressure; (2) high velocity and breaking impacts in a region of obstacles to allow the agglomerates to be broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and

c) mixing the dispersed clay solution with at least part of the rubber-modified pristine epoxy;

wherein the rubber of the rubber-modified pristine epoxy is CTBN, and a content of 6 phr of clay loading and 20 phr of CTBN yields the modified epoxy having, at about 20 phr of CTBN rubber and as low as about 4 phr clay loading, an increase in critical strain energy release rate ( $G_{IC}$ ) of at least 6 7.6 times, with respect compared to the rubber-modified pristine epoxy.

36. (Previously Presented) The modified epoxy of claim 35, wherein said step c) comprises adding curing agents to yield a solid epoxy material.

37. (Previously Presented) The modified epoxy of claim 20, wherein said step c) comprises adding curing agents to yield a solid epoxy material.



***Clean Version***

1. (Proposed Amendment) A method for making a modified epoxy, comprising the steps of:
  - a) preparing a clay solution comprising solvents, clay particles of a dimension in the nanometer range and agglomerates of clay particles;
  - b) generating a flow of clay solution and submitting said flow to: (1) high pressure to generate high velocity and to allow shearing in the clay solution to occur; (2) a region of obstacles allowing the agglomerates of clay particles to be broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and
  - c) mixing the dispersed clay solution with at least a pristine epoxy, wherein a rubber material is optionally provided with the pristine epoxy during mixing.
2. (Cancelled)
3. (Proposed Amendment) The method according to claim 1, wherein said step a) comprises incorporating a first part of the pristine epoxy into the clay solution; and said step c) comprises mixing the dispersed clay solution with a remaining part of the pristine epoxy.
4. (Original) The method according to claim 1, wherein said step a) comprises mixing with at least one of mechanical and ultrasonic mixing.

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5. (Original) The method according to claim 1, wherein said step b) comprises submitting the clay solution to a pressure of about 20,000 psi in tubes of a diameter of about 0.1 mm.
6. (Original) The method according to claim 1, wherein said step b) comprises exfoliating the clay particles in the solution.
7. (Previously Presented) The method according to claim 1, wherein said step c) comprises adding curing agents to yield a solid epoxy material.
8. (Proposed Amendment) The method according to claim 1, whereby the modified epoxy comprises agglomerates of less than about 1  $\mu\text{m}$  and agglomerates of a maximum diameter between about 1  $\mu\text{m}$  and 2  $\mu\text{m}$ .
9. (Previously Presented) The method according to claim 1, whereby the modified epoxy has enhanced viscoelastic properties and improved fracture toughness compared to the pristine epoxy.
10. (Proposed Amendment) The method according to claim 9, wherein a content of about 1 wt% of clay loading and no optional rubber yields an increase in critical stress intensity ( $K_{IC}$ ) and critical strain energy release rate ( $G_{IC}$ ) of 2 and 3 times respectively, with respect to the pristine epoxy.

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11. (Original) The method according to claim 1, whereby the modified epoxy has enhanced barrier properties, including water absorption resistance, adhesion strength and flammability resistance, with respect to the pristine epoxy.

12. (Original) The method according to claim 1, wherein a mixture of clay and epoxy obtained has a stability over an extended period of time.

13. (Original) The method according to claim 1, wherein the optional rubber is provided with the pristine epoxy during mixing.

14 - 19. (Cancelled)

20. (Proposed Amendment) A modified epoxy produced from a pristine epoxy, the modified epoxy having at least higher barrier properties and thermal resistance, a higher critical stress intensity factor ( $K_{IC}$ ), and a higher critical strain energy release rate ( $G_{IC}$ ) than the pristine epoxy, the modified epoxy produced by:

a) preparing a clay solution comprising solvents, clay particles of a dimension in the nanometer range and agglomerates of clay particles;

b) generating a flow of clay solution and submitting said flow to: (1) high pressure; (2) high velocity and breaking impacts in a region of obstacles to allow the agglomerates to be broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and

c) mixing the dispersed clay solution with at least part of the pristine epoxy, wherein a rubber material is optionally provided with the pristine epoxy during mixing;

wherein a content of about 1wt% of clay loading and no optional rubber yields an increase in  $K_{IC}$  and  $G_{IC}$  of 2 and 3 times respectively, with respect to the pristine epoxy.

21. (Original) The modified epoxy according to claim 20, comprising finely dispersed clay agglomerates of less than about 1  $\mu\text{m}$  and agglomerates of a maximum diameter between about 1  $\mu\text{m}$  and 2  $\mu\text{m}$ .

22. (Cancelled)

23. (Previously Presented) The modified epoxy according to claim 21, wherein the optional rubber is provided with the pristine epoxy during mixing.

24. (Original) The modified epoxy according to claim 21, further comprising additives.

25. (Cancelled)

26. (Proposed Amendment) The modified epoxy according to claim 23, wherein the optional rubber is CTBN, and a content of 6 phr of clay loading and 20 phr of CTBN yields an increase in  $K_{IC}$  and  $G_{IC}$  of 2.2 and 7.6 times respectively, with respect to the pristine epoxy.

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27. (Proposed Amendment) The method according to claim 13, wherein the optional rubber is CTBN, and a content of 6 phr of clay loading and 20 phr of CTBN yields an increase in critical stress intensity factor ( $K_{IC}$ ) and critical strain energy release rate ( $G_{IC}$ ) of 2.2 and 7.6 times respectively, with respect to the pristine epoxy.

28. (Proposed Cancellation)

29. (Proposed Cancellation)

30. (Proposed Cancellation)

31. (Proposed Cancellation)

32. (Proposed Cancellation)

33. (Proposed Amendment) A modified epoxy produced from a pristine epoxy, the modified epoxy having at least a higher flammability resistance and a higher fracture toughness than the pristine epoxy, the modified epoxy produced by:

a) preparing a clay solution comprising solvents, clay particles of a dimension in the nanometer range and agglomerates of clay particles;

b) generating a flow of clay solution and submitting said flow to: (1) high pressure; (2) high velocity and breaking impacts in a region of obstacles to allow the agglomerates to be broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine

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and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and

c) mixing the dispersed clay solution with at least part of the pristine epoxy;

wherein a content of 6 phr of clay loading yields an increase in critical strain energy release rate ( $G_{IC}$ ) of 5.8 times, with respect to the pristine epoxy.

34. (Proposed Amendment) The modified epoxy of claim 33, wherein said step c) comprises adding curing agents to yield a solid epoxy material.

35. (Proposed Amendment). A modified epoxy produced from a rubber-modified pristine epoxy, the modified epoxy having at least a higher flammability resistance and a higher fracture toughness than the rubber-modified pristine epoxy, the modified epoxy produced by:

a) preparing a clay solution comprising solvents, clay particles of a dimension in the nanometer range and agglomerates of clay particles;

b) generating a flow of clay solution and submitting said flow to: (1) high pressure; (2) high velocity and breaking impacts in a region of obstacles to allow the agglomerates to be broken down; and (3) a sudden lower pressure, yielding a dispersed clay solution having a fine and homogeneous distribution of clay particles of a dimension in the nanometer range in the clay solution; and

c) mixing the dispersed clay solution with at least part of the rubber-modified pristine epoxy;

wherein the rubber of the rubber-modified pristine epoxy is CTBN, and a content of 6 phr of clay loading and 20 phr of CTBN yields an increase in critical strain energy release rate ( $G_{IC}$ ) of 7.6 times, with respect to the pristine epoxy.

36. (Previously Presented) The modified epoxy of claim 35, wherein said step c) comprises adding curing agents to yield a solid epoxy material.

37. (Previously Presented) The modified epoxy of claim 20, wherein said step c) comprises adding curing agents to yield a solid epoxy material.

**Comments**

- The property limitations featured in claims 10 and 20 are supported by paragraphs 0091 and 0100 of the specification:

[0091] Nanocomposites obtained by the method of the present invention show a dramatic increase in fracture toughness at very low clay loading, with an increase in  $K_{IC}$  and  $G_{IC}$  of 2 and 3 times respectively at only 1.5-phr (about 1 wt %) organoclay loading.

[00100] In contrast, the method of the present invention enhances the degree of exfoliation of organoclay and breaks up agglomerates thereof. As a result, nanocomposites obtained with the method of the present invention show a dramatic improvement in fracture toughness at very low clay loading; that is,  $K_{IC}$  and  $G_{IC}$  are increased by 2 and 3 times respectively at 1.5-phr (about 1 wt%) organoclay loading over the pristine resin properties.

Note: the increase is by 2 and 3 times – not up to 2 and 3 times.

- The property limitations featured in claims 26 and 27 are supported by paragraphs 0092 and 0101 of the specification:

[0092] CTBN-modified nanocomposites, as compared to nanocomposites without rubber, show a further increase in both  $K_{IC}$  (Figure 23) and  $G_{IC}$  (Figure 24) (over rubber-modified epoxies) as the clay loading increases. All nanocomposites contain the same content of CTBN (20-phr) and different organoclay contents ranging from 0 to 6-phr. At clay loading of less than 3-phr, fracture toughness increases slowly, but, above this value, it dramatically improves.  $K_{IC}$  and  $G_{IC}$  are increased by 2.2 and 7.6 times respectively at 6-phr organoclay loading and 20-phr CTBN, compared with the pristine epoxy system. Therefore, there is a superposition effect on fracture toughness of hybrid epoxy nanocomposites modified with rubber and organoclay.



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[00181] In the case of rubber-modified epoxies, the present method further yields enhancement in the glass transition temperature  $T_g$  and mechanical performances. Modification with organoclay simultaneously improves the fracture toughness and compressive properties of DGEBA/ $\text{BF}_3\cdot\text{MEA}$ , that is,  $K_{IC}$  and  $G_{IC}$ , increased by 1.84 and 2.97 times, respectively; compressive modulus, ultimate strength, yield strength and fracture strain increased by 25.1 %, 29.1%, 5.8% and 9.6% respectively, at 6-phr concentration of CTBN, modification of the epoxy with organoclay and rubber not only further improves fracture toughness, that is,  $K_{IC}$  and  $G_{IC}$  are increased by 2.2 and 7.6 times respectively, at 8-phr organoclay loading and 20-phr CTBN compared to the pristine resin, but also enhances the glass transition temperature  $T_g$ , yield strength and ultimate strength compared with rubber-modified epoxies with a similar content of CTBN. Modification with organoclay improves the fracture toughness of TGDDM/DDS epoxy resin in which the strain energy release rate ( $G_{IC}$ ) of the virgin epoxy increases by 5.8 times with a clay loading of 6 phr.

Note: the increase is by 2.2 and 7.6 times – not up to 2.2 and 7.6 times.

- The property limitation featured in claim 33 is supported by paragraph 0093 of the specification:

[0093] Figure 25 is a graph of the critical strain energy release rate ( $G_{IC}$ ) of epoxy TGDDM/DDS obtained with a Direct Mixing Method (rhomboids) and with the method of the present invention (squares). It shows that the fracture toughness of the epoxy obtained by the present method is increases by 5.8 times the fracture toughness of the pristine epoxy.

- The property limitation featured in claim 35 is supported by paragraph 0092 of the specification:

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[0092] CTBN-modified nanocomposites, as compared to nanocomposites without rubber, show a further increase in both  $K_{IC}$  (Figure 23) and  $G_{IC}$  (Figure 24) (over rubber-modified epoxies) as the clay loading increases. All nanocomposites contain the same content of CTBN (20-phr) and different organoclay contents ranging from 0 to 6-phr. At clay loading of less than 3-phr, fracture toughness increases slowly, but, above this value, it dramatically improves.  $K_{IC}$  and  $G_{IC}$  are increased by 2.2 and 7.6 times respectively at 6-phr organoclay loading and 20-phr CTBN, compared with the pristine epoxy system. Therefore, there is a superposition effect on fracture toughness of hybrid epoxy nanocomposites modified with rubber and organoclay.